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STRUCTURE AND DYNAMICS OF EXCITED ATOMS

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by

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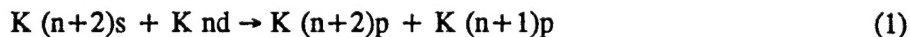
I. Introduction

During the five year period of grant AFOSR-90-0036 we have worked on two different kinds of projects, microwave multiphoton processes and resonant collisional energy transfer. Both kinds of experiments are carried out with highly excited, or Rydberg atoms, which allow us to do quantitative experiments giving insights which would be difficult or impossible to obtain in other systems. For example, our experiments on microwave multiphoton excitation and ionization have demonstrated that the Floquet, or dressed state, approach provides an accurate description of the evolution of atomic systems in intense radiation pulses. The understanding derived from our microwave experiments resolved a controversy about how laser multiphoton ionization through intermediate resonances occurred. In the two following sections of this report we describe the collision experiments and the microwave multiphoton experiments.

II. Resonant Collisional Energy Transfer

Resonant collisional energy transfer plays a crucial role in several laser media. For example, the HeNe laser works by resonant energy transfer from the excited He metastable 2s atoms to the Ne 3s states, the upper level of the 6328 Å laser transition.¹ A roughly analogous transfer from N₂ to CO₂ is important for the CO₂ laser.² In some solid state media the resonant dipole-dipole interaction between excited and ground state dopant ions leads to the production of two ions excited with half the energy, a process which converts near infrared light to mid infrared light with quantum efficiencies in excess of one.³ A quantum efficiency of two is possible.

We have studied intensively the resonant collision process⁴



which can be tuned into resonance with a small electric field. Here n is the principal quantum number. This process is a resonant dipole-dipole process and provides a system in which we can study resonant collisional energy transfer in astonishing detail. The collision cross section for the process of Eq. (1) is given by⁵

$$\tau = \frac{\mu_1 \mu_2}{v} \quad (2)$$

where μ_1 and μ_2 are the $(n+2)s - (n+2)p$ and $nd - (n+1)p$ dipole matrix elements and v is the collision velocity. The duration of the collision, the collision time T , is given by⁵

$$T = \sqrt{\mu_1 \mu_2} / v^{3/2} \quad (3)$$

For thermal atoms of principal quantum number $n = 25$ the cross section and collision time are 10^9Å^2 and 10^{-8}s . Inspecting Eq. (3) we can see that the duration of the collision increases rapidly with decreasing collision velocity, and this realization led us to velocity select the K atoms in a beam to reduce the collision velocity. We were able to observe collisional resonances as narrow as 1 MHz, and routinely 1.4 MHz wide, corresponding to collision times of $1 \mu\text{s}$.⁶ When we shortened the time between laser excitation and field ionization detection below $1 \mu\text{s}$ we observed the linewidths of the collisional

resonances to broaden. In this case the collisional resonances are transform limited. Furthermore, we know exactly when individual collisions begin and end. To our knowledge this has never been possible previously with a quantum system. This ability allows us to perturb the colliding atoms at well defined points in the collision, for example, when the atoms are at their point of closest approach.

We have used the above capability to study radiative collisions, collisions in the presence of a radiation field in the regime in which the radiation frequency ν is the inverse of the collision time T .⁷ In this regime it is apparent that the phase of the radiation field is likely to be important. Prior to our experiments this regime had not been considered at all; only the high and low frequency regimes $\nu \gg 1/T$ and $\nu \ll 1/T$ had been considered. We found that with a phase locked microwave field the atomic levels of the two colliding atoms could be brought into resonance twice during the collision, and interference fringes due to the phase accumulation between the two times at resonance were plainly visible. This description of the observations is based on an inherently low frequency description, i.e. the evolution during each field cycle is treated explicitly by computing transition amplitudes each time the states come into resonance using the Landau-Zener theory.⁷

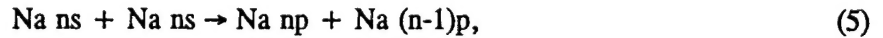
Equally as interesting as the fact that we can observe and understand collisions with $\nu \approx 1/T$ is the fact that we have shown how to connect these observations to the high frequency regime, which is based on a Floquet, or dressed state approach. The connection has been made both in an approximate way which gives excellent physical insight and more formally to show that the connection can be made rigorously.^{7,8}

Having the ability to observe 1 MHz wide collisions using the system of Eq. (1) we carried out extensive experiments over the whole frequency range $\nu \ll 1/T \ll \nu$. These measurements verified aspects of the theory of radiative collisions in strong fields which had not previously been examined and showed that the theory was in fact correct.⁹ For example, collisions in which m rf photons are absorbed or emitted during the collision should have cross sections given by¹⁰

$$\sigma_m = \sigma_0 \left| J_m \left(\frac{KE_{rf}}{\omega} \right) \right|, \quad (4)$$

where σ_0 is the cross section without rf, k is the permanent dipole moment of the nd state in the static field, J_m is a Bessel function, and ω and E_{rf} are the angular frequency and amplitude of the rf field. We have shown experimentally that the Bessel function description of Eq. (4) is correct.

While most of our efforts have been focussed on the K process of Eq. (1), we have also examined the Na process¹¹



which is also tuned into resonance with an electric field. All the K measurements above and all the previous Na measurements had been done in purely electric fields. In Na the spin is decoupled from the orbital angular momentum, and our presumption was that the spin was unchanged in the collisions. To test this notion we have done experiments with \vec{B} as well as \vec{E} fields to lift the degeneracy of the spin states.¹¹ With $\vec{B} \parallel \vec{E}$ it is apparent that there are either no spin flips, or the two spins flip in opposite senses in the two colliding atoms. Since the latter seems unlikely, we can safely conclude that collisions do not alter the spins of the two colliding atoms.

III. Microwave ionization and excitation

In multiphoton ionization by visible light there is a pronounced difference between ionization by circularly and linearly polarized light.¹² It requires higher intensity with circular polarization and the differences between linear and circular polarization increases with the order of the process. Since ionization of $n = 25$ atoms by microwaves requires hundreds of photons we expected to see a huge difference between linear and circular polarizations. In fact, we did. Ionization by a linearly polarized field requires a field amplitude¹³

$$E_{tn} = 1/3n^5, \quad (5)$$

while ionization by a circularly polarized field requires a field¹⁴

$$E_{\text{clrc}} = 1/16n^4, \quad (6)$$

essentially the same requirement as for ionization by a static field. The similarity of the requirement of Eq. (6) to the static field is not so surprising since in a frame rotating with the microwave field, it is in fact static. Any deviation from circular polarization at $E \sim 1/16n^4$ leads to very rapid ionization, for then the field is not static in the rotating frame and can drive transitions, allowing ionization at lower fields. Ionization by circularly polarized fields has attracted much theoretical interest, and to date our measurements are the only ones published.

The understanding derived from the ionization measurements made it clear that it should be possible to produce circular states, with $|m| = \ell = n - 1$ using microwave fields, and during this period we demonstrated a technique which is based on exciting a Na $(n+1)s$ state in a circularly polarized microwave field then reducing the field to zero. En route the atoms make an adiabatic rapid passage through the $n - 1$ photon resonance with the $\pm m = n - 1$ state and are left in the circular state. The choice of $m = \pm (n - 1)$ is determined by the sense of the field rotation.

The fact that slight deviations from circular polarizations lead to ionization led us to examine ionization in combined static and microwave fields an approximately equivalent situation.¹⁵ These measurements showed that in strong static fields, $E > 1/3n^5$ where there are numerous level avoided crossings that the added rf field must be enough to sample the nearest level crossing, a reasonable result. The most striking result was that for static fields above $E = 1/6n^5$ raising the static field raised the microwave field required, a surprising result. The reason is as follows. If we think of ionization as transitions among static Stark states, for ionization to occur the total field must sample avoided crossings to go to higher lying states. At zero field the avoided crossing from $n \rightarrow n + 1$ at $E = 1/3n^5$ and the zero field $\Delta n = 0$ avoided crossings are both important. When the static field exceeds $E = 1/6n^5$ it is

easier to reach the $n \rightarrow n+1$ avoided crossings than those at zero field, and they are the rate limiting step, so that for $1/6n^5 < E_{\text{static}} < 1/3n^5$ the required microwave field is

$$E_{\text{mw}} = E_s. \quad (7)$$

One of the more surprising features of multiphoton ionization is above threshold ionization, ATI, the absorption of more photons than needed for ionization. In the prior grant period we observed ATI using microwave fields and developed a simple classical picture to describe our observation.¹⁶ Equivalent pictures were developed by others, and this classical picture is sometimes called the Simpleman's picture. This model leads immediately to several interesting predictions. First, it implies that if the phase at which ionization is known the energy of the ejected electron can be controlled. We have demonstrated this point clearly by phase locking the microwaves to a picosecond laser and selecting the phase.¹⁷ As predicted, if the electrons are ejected at a zero crossing of the microwave field the observed electrons have energy $3U_p$ while if they are ejected at the field maximum they have energy U_p . Here U_p is the ponderomotive energy, $U_p = \frac{E^2}{4\omega^2}$, the energy of a free electron oscillating in the field $E \cos \omega t$.

The Simpleman's model also predicts that with circularly polarized light electrons of energy $2U_p$ are observed, in the plane of polarization. More surprising, with slightly elliptical polarization, made of two orthogonal polarizations 90° out of phase, the electrons will be ejected in the direction of the weaker component, because ionization occurs at the instant the stronger component reaches its maximum, in which case $3/4$ of the ATI energy comes from the weaker component.¹⁸

We have studied two aspects of ionization by linearly polarized microwaves. As shown by Eq. (5) ionization occurs at $E = 1/3n^5$. We originally asserted that it was via a sequence of Landau-Zener transitions through higher lying states culminating in ionization, with the rate limiting step being the $n \rightarrow n+1$ transition requiring a field $E = 1/3n^5$. Our later measurements of K multiphoton transitions suggested that this picture might not be completely correct, the $n \rightarrow n+1$ transitions might be resonant. We have made measurements of ionization of Na at frequencies of 10 MHz to 30 GHz, and there are too

few field cycles at the lowest frequencies for the Landau-Zener picture to be possible.¹⁹ The higher efficiency of a resonant process is absolutely necessary. Recently we have begun to study another aspect of the ionization problem. Many intermediate states must be traversed en route to ionization, and with short pulses atoms might be left in states higher than the initial state. Surprisingly, even with pulses as short as 3ns, 25 cycles at 8 GHz, we see very few atoms left in states other than those initially populated.²⁰ We have recently generated 8 GHz pulses as short as 500 ps, and we look forward to observing ionization with them.

Much of our effort has been focussed on multiphoton transitions in K, specifically in the two level system consisting of the $(n+2)s$ state and the lowest $n,3$ state of the adjacent n Stark manifold. The Stark state n,ℓ is adiabatically connected to the zero field $n\ell$ state. We had previously worked out a Floquet theory for the multiphoton Rabi frequencies between the K $(n+2)s$ and $n,3$ Stark states, which predicted that the Rabi frequency, ω_m , of the m photon transition was given by²¹

$$\omega_m = \omega_0 J_m (kE_{mw}/\omega), \quad (8)$$

where ω_0 is the size of the static field anticrossing, J_m is a Bessel function, E_{mw} and ω are the microwave field amplitude and angular frequency, and k is the dipole moment of the Stark state. In this grant period we explicitly measured the Rabi frequencies and verified the Bessel function dependence of Eq. (8), including the fact that the Rabi frequencies exhibit the zeros of the Bessel functions²²

These results demonstrate that our description of the multiphoton processes is correct, so that we are sure that we understand the atoms in a continuous wave microwave field. We then exposed the atoms to pulses of microwaves short enough that the field inhomogeneties did not limit the resolution. With these pulses we were able to show that the evolution of the atomic system could be described as the evolution of the Floquet, or dressed, levels, during the microwave pulse. Specifically, we examined the K 21s - 19,3 4 photon 9.3 GHz transition in static fields of approximately 235 V/cm.²³ The AC Stark shift due to the microwave pulse brought the two levels into resonance on the rising and falling edges of

the pulse. At the resonance the two Floquet levels have an avoided crossing, equal to the Rabi frequency, and how the avoided crossing is traversed depends on how rapidly it is traversed relative to its own size. Our experiments showed clearly that how the crossing is traversed can be predicted exactly using the Landau-Zener theory. The most dramatic illustration of this point is that for intermediate rates of traversal the probability amplitude splits, with part following each state, on the first traversal of the resonance and is recombined on the second, leading to quite evident interference fringes. To our knowledge this experiment was the first demonstration that the evolution of the Floquet states provided the key to understanding how atoms respond to intense radiation pulses. Applying this understanding to laser multiphoton processes later resolved some of the confusion surrounding those measurements.^{23,24}

IV. Conclusion

This report is a summary of the research done under grant AFOSR-90-0036. A quantitative study of the effects of strong microwave fields on Rydberg atoms, research which has already had an impact on laser multiphoton research. The second, resonant collisional energy transfer between Rydberg atoms, represents the first time anyone has been able to know when individual collisions ability to study experimentally radiatively assisted collisions with electromagnetic fields phase locked to the collisions.

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